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Chemical and Biological Detectors using Ultra-High-Q Microresonators

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Recently, a method for fabricating planar arrays of optical microtoroid resonators with quality factors greater than 500 million was developed. These devices have previously demonstrated Raman and OPO lasing and radiation pressure induced oscillations. When immersed in an aqueous environment, these devices are able to maintain their ultra-high Q factors by operating in the visible wavelength band, enabling very sensitive chemical and biological detection. The fabrication and optical properties of these devices will be described. These devices have performed both chemical and biological detection. Systems which have been detected include D_2O in water and a variety of biological molecules. Sensitivity limits will also be discussed.

High Q and Ultra-high Q (UHQ) silica optical microcavities can perform as highly sensitive detectors [1-3]; their excellent transduction abilities is a result of the long photon lifetime within the whispering gallery of the microcavity. This signal amplification which is inherent to the device is unique among optical sensors. For example, in a waveguide sensor the photon interacts with the functionalized surface only once [4]; whereas the recirculation within the microcavity allows photons to interact with the surface many times, thus amplifying a single detection event. Additionally, the surface of silica-based microcavities is easily sensitized using silanization agents [5], amines, carbohydrates, the biotin-streptavidin system [6] or antibodies [7]. For example, silica microsphere resonators, with a properly sensitized surface, were recently used to distinguish between two strands of DNA [2]. Because the sensitivity increases as the quality factor increases, it is very important to maximize the Q of the cavity. In these previous experiments, the Q was limited to 1 million (10^6) which is over 100 times lower than in air. If the Q is increased to above 100 million, then label-free single molecule detection would be theoretically possible.[2]

There are many loss mechanisms which contribute to the quality factor of a resonant cavity, such as material loss, radiation loss, surface roughness loss, contamination loss and coupling loss.[8] While detailed studies have been performed to determine the limits of a resonator's quality factor in air [8], [9], [10], no comparable studies have been performed in water. Because most biological detection experiments are performed in a water-based solution, it is important to thoroughly understand the impact of this environment on resonator and optimize all parameters of the system (operational wavelength, resonator size, resonator material) to achieve the highest possible quality factor. By maximizing the Q of the cavity, the sensitivity of the cavity will also be optimized.

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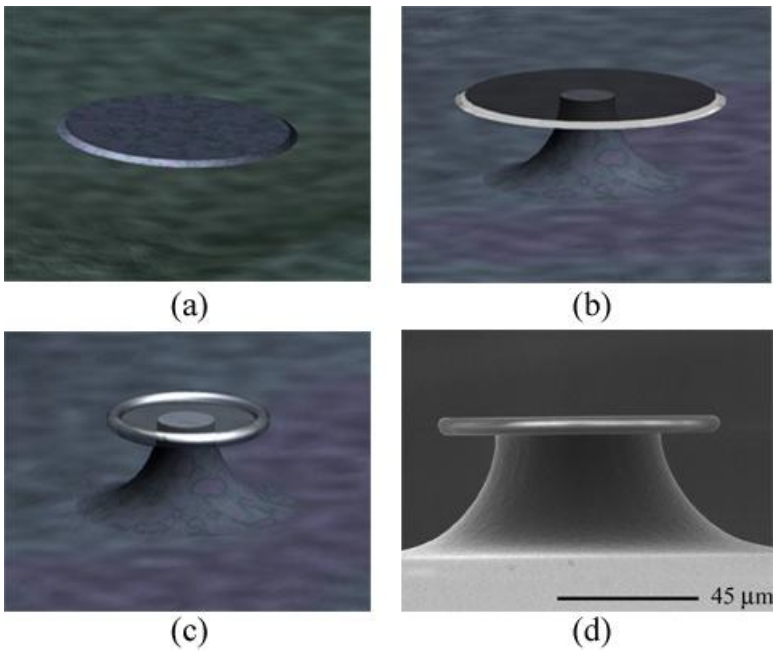

Figure 1: The fabrication of the ultra-high-Q microtoroid resonator consists of a single three step process. a) patterning the silica into an array of circular oxide pads, b) etching the silicon wafer substrate with XeF_2 (an isotropic, gas-phase etchant) to form the high-Q microdisk, and c) reflowing the silica microdisk with a CO_2 laser to form the toroidal resonator. d) Scanning electron micrograph of the UHQ microtoroid resonator.

To this end, UHQ silica microtoroid resonators were fabricated over a wide range (50-250micron) of major toroid diameters using a previously outlined process (Figure 1). [11] Experiments were performed in both water (H_2O) and deuterium oxide (D_2O). The D_2O was purchased from Aldrich. D_2O was chosen as the second liquid because it has the same refractive index and, in turn, the same radiation-loss as H_2O. However, its absorption at all wavelengths tested is significantly less.[12] This contrast allowed for selective probing of the absorption-loss mechanism and verification of the model developed to describe this system.[13]

The model uses finite-element-analysis to predict the Q factor of microtoroid resonators immersed in water and accounted for two loss mechanisms: radiation-loss and absorption-loss. Both mechanisms are modified for aqueous versus air operation. In particular, the reduced refractive-index contrast for aqueous operation increases radiation loss at a fixed resonator radius, while water or D_2O make absorption within the environment the central factor in limiting Q at large radii. It is important to note that the refractive index of water and D_2O are identical, while the absorption of D_2O is significantly less than water. This results in identical radiation-loss quality factors for both liquids but significantly higher absorption-loss quality factors for D_2O.

Measurements of the resonator quality factor and analysis of the modal structure were performed at three wavelength bands (680, 1300 and 1500 nm). By using these three wavelengths, a large range of the optical spectrum could be probed. Additionally, a large variation in optical absorption of either heavy water or water could be verified. For testing, a

single-mode, tunable external cavity laser was coupled to a single-mode optical fiber containing a short, tapered section. Tapered fiber waveguides are high-efficiency probes of microcavities [14, 15]. The tapered-fiber waveguides are fabricated by heating an optical fiber using an oxyhydric torch, while stretching the fiber. Tapered fibers for testing at 680nm were pulled from F-SV fiber to an average waist diameter of 500nm. Tapers for testing at 1330nm and 1550nm were pulled from SMF fiber to an average diameter of 1 micron. The tapered section was used to couple power into the "whispering gallery modes" of the UHQ microtoroids.

During testing, the UHQ microtoroids were placed on a high-resolution translation stage (100-nm step resolution) and were monitored by two cameras (top and side view) simultaneously. The quality factor of the microtoroid resonator was first determined in air to ensure that it was above the theoretical limit for a given toroid diameter (once immersed in liquid). Then, with the taper waveguide in close proximity to the microtoroid, liquid was added and a cover slip was placed on top (Fig. 2). A "liquid" gap between the toroid and the taper was maintained when determining the quality factor in either H_2O or D_2O in order to maintain constant coupling between the microtoroid resonator and the taper waveguide.

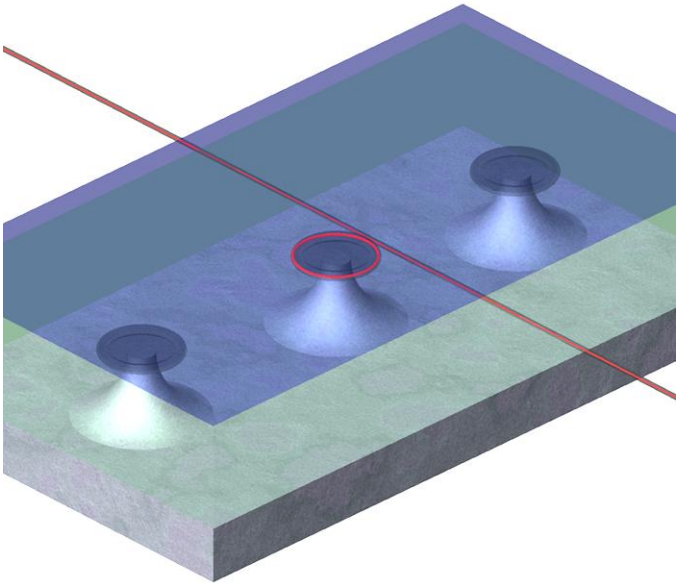

Figure 2. The Ultra-High-Q silica microtoroid is first coupled to the fiber taper waveguide and the Q is determined in air. After being immersed in either H_2O or D_2O, a cover slip is placed on top forming a microaquarium.

Figure 3 shows typical transmission spectra in H_2O at 1300nm and in D_2O at 1550 nm. The spectra are taken in the under-coupled regime [14]. The modal structure is dominated by principal transmission minima, confirmed below, to be the fundamental transverse mode of the microtoroids. The intrinsic, Q factor (i.e., the Q factor in the absence of waveguide loading) was determined by scanning the single-mode laser (short-term linewidth of 300 kHz) and measuring both the transmission and the loaded linewidth (full-width-half-maximum) for several waveguide-resonator coupling conditions in the under-coupled regime.

The intrinsic modal linewidth (and hence intrinsic Q) was then computed using a simple coupling model.[14] The laser scan frequency was optimized so as to ensure that neither scan direction (increasing frequency vs. decreasing frequency) nor scan frequency had any observable impact on linewidth.

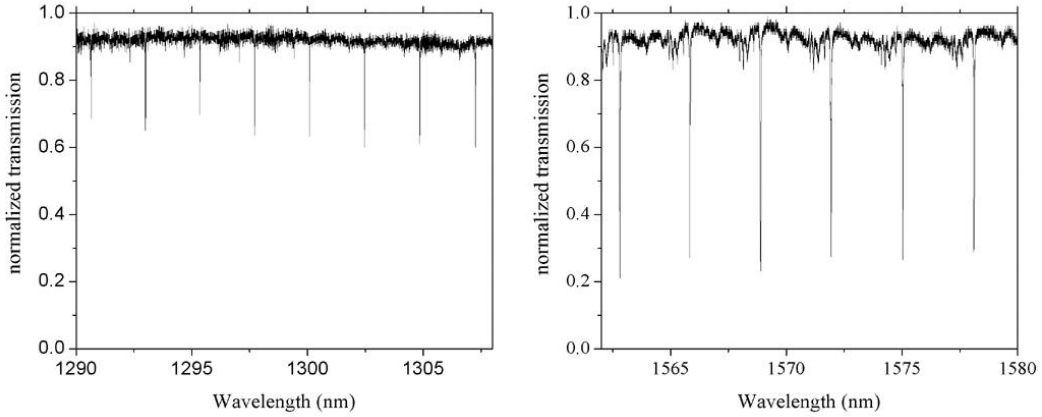

Figure 3. Transmission spectra of a microtoroid resonator in D_2O at 1300nm and 1550nm bands. The resonator is highly under-coupled in the 1300nm spectra. In the 1550nm spectra, the resonator is also under-coupled but closer to being critically coupled.

The intrinsic Q factors measured in the 680 nm band plotted versus toroid major diameter are presented in Fig. 4 (triangular and circular points). Q factors trend to larger values with increasing toroid size. This behavior is in good agreement with predictions of the model (also shown in Fig. 4) and results from radiation loss. The maximum quality factor achieved in H_2O was 2.3×10^8 and in D_2O was 1.3×10^8. These values are notable as they represent the highest Q factors reported to date for operation in an aqueous environment. Measurements beyond Q factors of 500 million were not possible in this experiment owing to laser linewidth stability. In principle, however, larger toroid diameters should exhibit quality factors as high as 1×10^9, in water, and 1×10^{10} in D_2O.

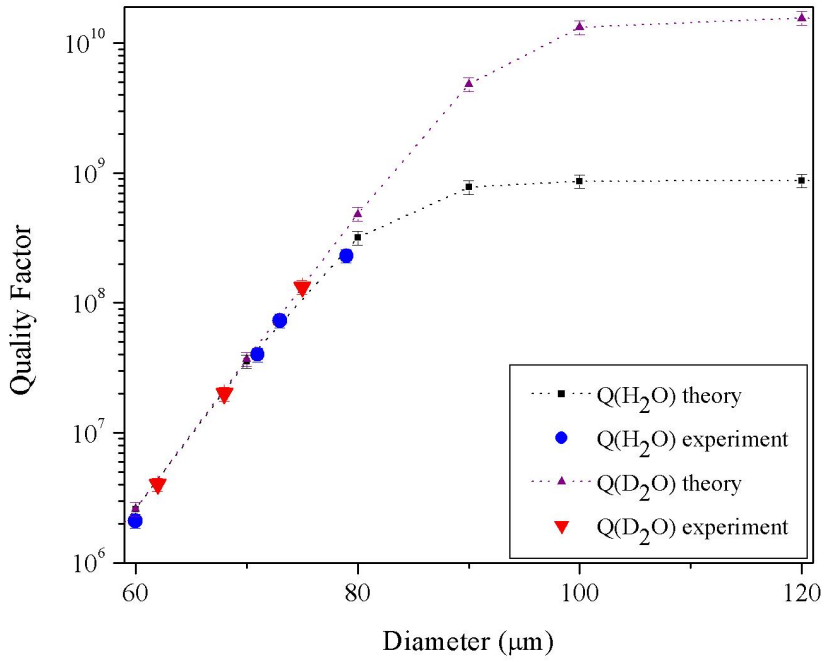

Figure 4. Quality factors measured and predicted in the 680nm band plotted versus toroid major diameter. Q increases with major diameter over the range of diameters wherein radiation loss is the dominant loss mechanism. It then plateaus at values set by absorption of the aqueous environment. Above $5x10^8$ data taking is unreliable due to laser-linewidth stability limitations. The maximum quality factor achieved in H_2O was $2.3x10^8$ and in D_2O was $1.3x10^8$.

The measured intrinsic Q factors for microtoroids in H_2O and D_2O at different toroid diameters and measured in the 1300nm band are plotted in Fig. 5. Both the radiation-loss-limited regimes and the absorption-loss-limited regimes are clearly visible in these plots. Also plotted are predictions based on the model. Within this wavelength band, D_2O has a lower optical absorption and hence exhibits an absorption-limited Q plateau that is significantly higher than for H_2O (approximately 10^6 for H_2O versus above 10^7 for D_2O). The origin of this absorption limit is the vibration overtone of water. In D_2O this overtone is wavelength-shifted significantly, thereby increasing the observable Q plateau.

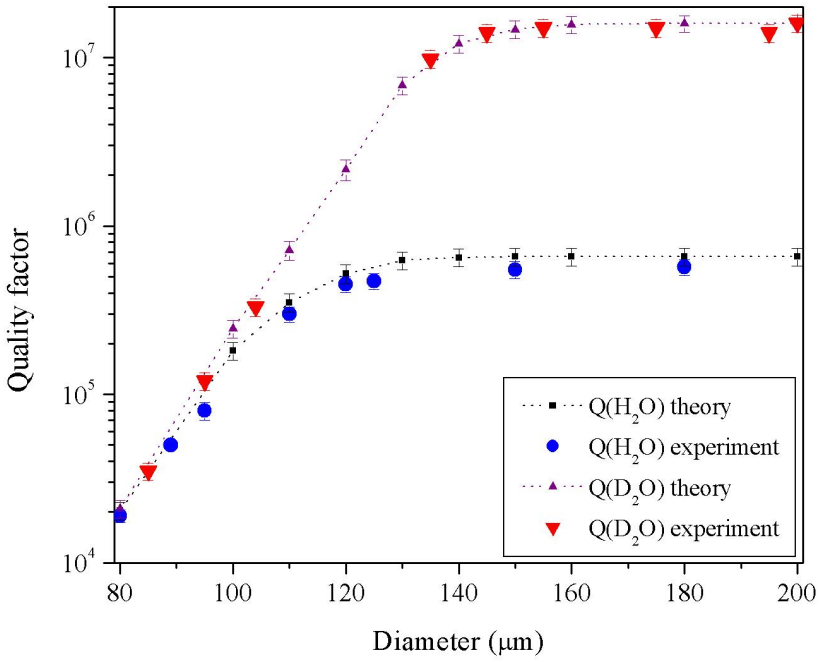

Figure 5. Quality factors measured and predicted in the 1300nm wavelength band. Both the radiation-loss-limited (small toroid diameter) and aqueous-absorption-loss limited regimes (Q plateau) are apparent. The measured absorptive-loss limits are $5x10^5$ (in H_2O) and $1.6x10^7$ (in D_2O).

The measured intrinsic Q factors versus toroid diameter in the 1550nm band are shown in Fig. 6, along with the predictions of the model. Again, there is good agreement between measurement and the model, showing the transition between the radiation-loss-limited and absorptive-loss-limited regimes. The strong OH overtone absorption in H_2O lowers the Q plateau to $8x10^4$, while for D_2O the value is higher, increasing to above $3x10^6$.

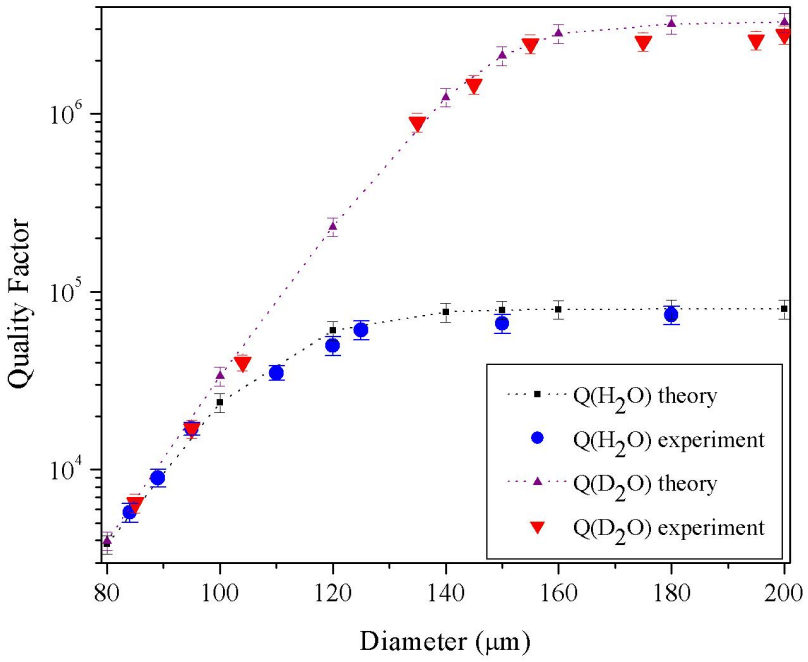

Figure 6. Quality factors measured and predicted in the 1550nm band. In H_2O, the maximum quality factor achieved is $7x10^4$. By changing to D_2O, the maximum quality factors increased to $2.8x10^6$.

Having achieved one of the requirements for a highly sensitive resonant cavity sensor (ultra-high-Q in water), a very simple yet elegant demonstration of this sensor's detection abilities presented itself in the form of heavy water detection.[16] At 1300nm, there is a large difference in the optical absorption of heavy water and water. This difference leads to a large change in the cavity Q depending on the percentage of heavy water in the environment around the toroid. Therefore, by monitoring the Q, it is possible to determine how much heavy water is present in the water.

To demonstrate this effect, a simple testing procedure was designed: 1) immerse the microtoroid in 100% D_2O, 2) gradually increase the concentration of H_2O in D_2O, until 100% H_2O is reached, and 3) return the concentration of D_2O to 100%[16]. The difference between the quality factor in H_2O and D_2O is liquid-limited[13]; therefore, the quality factor can be described by: $Q_{liq}=2\pi n/\lambda\alpha$, where n=effective refractive index, λ=wavelength, and α is the absorption loss due to the liquid. The refractive index of H_2O and D_2O are similar and the resonant wavelength is constant.

During the initial series of measurements, the solutions were prepared in 10% increments (10% H_2O in D_2O, 20% H_2O in D_2O, etc), starting with 100% D_2O. After the quality factor was determined, all of the D_2O was removed, and the chamber was then flushed five times with the next concentration solution (in this case, the 10% H_2O in D_2O), and the quality

factor was again determined. This flushing process was followed for all solutions to remove trace amounts of higher or lower concentration solutions. As can be seen in Figure 7a, when the concentration of D_2O was reduced, the quality factor decreased. The theoretical values for each concentration were calculated and are indicated by the dashed line. This Q decrease was reversible, and by increasing the D_2O concentration, the quality factor is recovered.

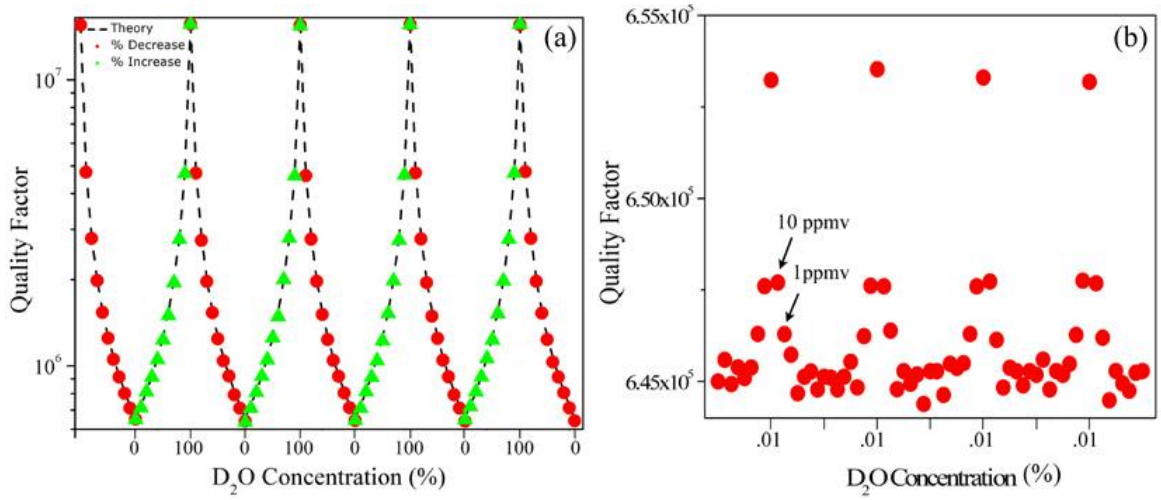

Figure. 7: a) The Q is systematically degraded (circles) and recovered (triangles) as the D_2O and H_2O are exchanged repeatedly. b) Starting with 100% H_2O, the concentration of D_2O was gradually increased using low concentration solutions ranging from $1x10^{-9}$% to .01%. The minimum detectable change in Q was at .0001% (1ppmv).

To determine the lower bound of the detection limit, larger dilutions of D_2O in H_2O were prepared, ranging from .01% to $1x10^{-9}$%. As can be seen in Figure 7b, there is a strong signal at .001% D_2O in H_2O and a small, yet detectable, shift occurs with the .0001% D_2O solution. These values are not believed to reflect the fundamental limit of the detection sensitivity of this device since no attempt was made to reduce operational sources of noise.

The ultra-high-Q microcavity has demonstrated the ability to detect the difference between two chemically similar species, H_2O and D_2O, at low concentrations. This detection is based on the subtle difference in optical absorptions between D_2O and H_2O, which is then magnified by the quality factor of the resonator. Using resonant cavities, it was possible to improve upon previous detection sensitivities (30ppmv) by over an order of magnitude.

In biological detection experiments, both specificity and sensitivity are important. While the ultra-high-Q optical resonator is inherently sensitive, as was demonstrated by the resonator's Q values > 10^8, specificity is achieved by functionalizing the surface of the microtoroid. Several different surface functionalization techniques have been used (antibody, biotin), each one specific to the target molecule.

Both the biological detection and the heavy water detection experiments using the ultra-high-Q toroidal resonators will be presented. Additionally, fundamental sensitivity limits will be discussed.

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